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METHOD OF ESTIMATING THE LEAKAGE OF MULTIPLE BARRIERS IN A RADIOACTIVE MATERIALS SHIPPING PACKAGE

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ABSTRACT

This paper presents the results of a theoretical study of the performance of multiple leaky barriers in containing radioactive materials in a shipping package. The methods used are reasoned analysis and finite element modeling of multiple barriers. The finite element model is developed and evaluated with parameters set to bracket 6M configurations with three to six nested plastic jars, food-pack cans, and plastic bags inside Department of Transportation (DOT) Specification 2R inner containers with pipe thread closures. The results show that nested barriers reach the regulatory limit of 1×10^{-6} A_2/hr in 11 to 52 days, even though individually the barriers would exceed the regulatory limit by a factoı of as much as 370 instantaneously. These times are within normal shipping times. The finite element model is conservative because it does not consider the deposition and sticking of the leaking radioactive material on the surfaces inside each boundary.

BACKGROUND

Transportation safety regulations require that the containment boundary of a type B packaging assure that the leakage of radioactive material is no greater than 1×10^{-6} A_2/hr under Normal Transport Conditions and 1 A_2 in a week under Hypothetical Accident Conditions. This paper is part of a continuing study of the efficacy of multiple barriers for transportation packaging.

SHIPPING PACKAGE CONFIGURATION

The DOT Specification 6M shipping package configuration is used in this study. It consists of nested food-pack cans and plastic bags inside a schedule 40, 5-inch pipe closed with threaded caps, which is, in turn, centered inside an open-headed drum with cane fiberboard.

This package has performed well in maintaining adequate containment of radioactive materials, including plutonium oxide. The method presented in this paper predicts a radioactivity leakage rate through multiple barriers that is much smaller than predicted from the gas leakage rate of the barriers taken individually. The better performance of the multiple barriers over a single barrier with the same gas leakage rate arises from the fact that, although the radioactivity leakage rate of a single barrier is a simple multiple of the gas leakage rate, the radioactivity leakage rate of multiple barriers is affected by dilution and deposition inside each barrier and, therefore, is dependent on time as well as the gas leakage rate.

A brief review of the relationship between radioactivity leakage rate and gas leakage rate of a single barrier will help in understanding the effect of time in the multiple barriers. Before a package is shipped, the leakage rate across the containment boundary (a single barrier) is measured using a tracer gas, usually helium. The radioactivity leakage rate is calculated from the measured gas leak rate using the methods specified in N14.5 of the American National Standards Institute. This paper considers shipments of plutonium oxide for which the material available for leaking is air containing aerosol particles of plutonium oxide. There are three numerical values for the leakage rate as follows:

1. The leakage rate of the tracer gas (helium) in the leakage test.

2. The leakage rate of the carrier gas (air), which is determined from the leakage rate of the tracer gas by the methods of N14.5.

3. The leakage rate of the radioactive material, which is the leakage rate of the carrier gas times the density of the radioactive aerosol particles in the carrier gas as given in N14.5.

At this point we must make it clear that the multiple barriers this paper is examining are not the same as the double-containment boundaries that are required by the regulations for shipping plutonium oxide.

The gas leakage rates of the individual food-pack cans, plastic bags, and the pipe thread closure of the 2R inner container that enclose the plutonium in a 6M package are all credited with a gas leakage rate of 1×10^{-3} std cc/sec air, based on the fact these components are all demonstrated to be watertight by gas-bubble leak testing. Table A1 of ANSI N14.5 credits the various gas-bubble leak tests with a sensitivity of 10^{-2} to 10^{-4} std cc/sec air with 1×10^{-3} std cc/sec air being the appropriate value for the conditions under which the gas-bubble leak tests are done with the 6M shipments. That a gas-bubble leak test corresponds to watertight is obvious from the fact that if gas bubbles do not form when the component is submerged in water, water is not getting in.

The radioactivity leakage rate of an aerosol of plutonium oxide particles through a single barrier with a gas leakage rate of 1×10^{-3} std cc/sec air is obtained using the aerosol density value provided in Appendix B16.33 of N14.5 of 9×10^{-6} gm/cc of the carrier gas. A gas leakage rate of 1×10^{-3} std cc/sec air corresponds to a radioactivity leakage rate of 3.7×10^{-4} A_2/hr plutonium, which is 370 times the regulatory limit of 1×10^{-6} A_2/hr.

Although multiple barriers have not been analytically studied as a possible way of satisfying transportation regulations, they are commonly used to contain radioactive material at all nuclear facilities. Radioactive material is double- or even triple-bagged when it is removed from glove boxes. Radioactive material is frequently stored in nested metal and plastic cans and jars. There is historical precedent, therefore, for the use of multiple barriers in containing radioactive material, although the authors are not aware of any previous study of why multiple barriers are successful.

FUNDAMENTALS OF LEAKING

There are two elements involved in leaking:

1. There must be one or more leak paths.
2. There must be a driving force to cause flow through the leak paths.

For transportation packaging, gas leak tests are performed to determine if leak paths are present and to determine if the radioactivity leak rate is within regulatory limits for the package contents.

There are several possible driving forces for transportation packaging. Decay heating of the contents will pressurize the inside of a package's containment boundary, but once thermal equilibrium is reached and the pressure is relieved, it would no longer cause leakage. In addition to decay heating, there are the daily temperature changes, weather changes in temperature and atmospheric pressure, and pressure changes due to elevation changes during travel. These other driving forces cause flow both in and out of the leak path. The Nuclear Regulatory Commission (NRC) makes the conservative assumption that there is always a driving force to cause flow out through a leak path. Because this paper is attempting to evaluate compliance with the NRC regulatory limit, the NRC's methods are followed. In other words, this paper assumes a constant driving force to cause leakage.

Given a constant driving force, a single barrier operates at a constant gas leakage rate. If the barrier's corresponding radioactivity leakage rate exceeds 1×10^{-6} A_2/hr, the barrier does not comply with the regulatory limit.

Finite Element Model

Two Barriers. What does nesting two or more barriers that individually exceed the regulatory limit on radioactivity leakage accomplish? This paper addresses this question by means of a finite element model of nested barriers. The model solves eight simultaneous linear differential equations for each barrier; five equations for the mass balance of the carrier gas and three equations for the mass balance of radioactive aerosol particles. The method of solution used with this paper is a matrix of finite element equations. Mathcad 4.0 is the program used to solve the matrix.

For the carrier gas mass balance in the finite element model, the carrier gas is considered to behave as a perfect gas and the leak hole diameter in each barrier is considered to be related to the values measured in the leak test by Eqs. B2, B3, and B4 of ANSI N14.2. The size of the leak hole in each barrier is held constant at the value measured in the gas leak test, which is performed with a 1-atmosphere pressure drop across the barrier. Because the gas leakage test is performed with a 1-atmosphere pressure drop across the barrier, the model sets the pressure inside the first or source barrier at 2 atmospheres and the initial pressure inside downstream barriers at 1 atmosphere. Because the size of the leak hole does not change, the pressure within the free space inside each barrier except the source barrier rises until the pressure drop from the source barrier to ambient is equally divided across all the barriers. This results in the gas leakage rate of each barrier being reduced when they are nested together.

The following matrix is the finite element form for the mass balance of the radioactive aerosol particles in the gas space between two nested barriers. The first barrier is the infinite source of radioactive particles, and it is nested within the second barrier. Equations (1) through (5) are for the mass balance of the carrier gas inside the second barrier, and Eqs. (6) through (8) are for the mass balance of the radioactive aerosol particles inside the second barrier. Equation (9) sums the mass of radioactive aerosol particles that pass through the second barrier to the ambient. The following paragraphs describe the terms and equations in the matrix.

The barriers are identified by the lowercase letters:

"a" for the source barrier,

"b" for the second barrier, and

"amb(ient)" for everything outside the second barrier.

(With up to six barriers, the additional barriers are identified by letters "c" . . . "f." Everything outside the last barrier is identified as "amb(ient).")

$$\left[\begin{array}{lll}
La_{i+1} & = \left[\dfrac{Fm}{\dfrac{Pa+Pb_i}{2}}+Fc\right][Pa-Pb_i] & (1)\\
Lb_{i+1} & = \left[\dfrac{Fm}{\dfrac{Pb_i+Pamb}{2}}+Fc\right][Pb_i-Pamb] & (2)\\
delNRTb_{i+1} & = [(La_i*delt*Pb_i)-(Lb_i*delt*Pb_i)] & (3)\\
sumNRTb_{i+1} & = [sumNRTb_i+delNRTb_i] & (4)\\
Pb_{i+1} & = \left[\dfrac{sumNRTb_i}{Vb}\right] & (5)\\
ADb_{i+1} & = if\left[\left(\dfrac{sumRMb_i}{Vb}\right) < AD\ ,\left(\dfrac{sumRMb_i}{Vb}\right)\ ,\ AD\right] & (6)\\
delRMb_{i+1} & = [(La_i*delt*AD)-(Lb_i*delt*ADb_i)] & (7)\\
sumRMb_i & = [sumRMb_i+delRMb_i] & (8)\\
sumRMamb_{i+1} & = [(sumRMamb_i)-(LB_i*delt*ADb_i)] & (9)
\end{array}\right]$$

Equations (1) and (2) are the carrier gas leakage rates (La, Lb) from Eq. B2 of ANSI N14.5. Fm and Fc are from Eqs. B3 and B4 of ANSI N14.5 and are evaluated at the leak hole diameter inferred from the gas leak test. P is the pressure within the designated barrier in atmospheres.

Equation (3) sets the mass of carrier gas that stays within the second barrier in a finite time element equal to the carrier gas leaking in, minus the carrier gas leaking out in the preceding finite time element. The terms on the righthand side of Eq. (3) are the carrier gas leakage rates for the first and second barriers, La and Lb, in cc/sec multiplied by the size of the finite element, delt, in sec multiplied by the pressure, P, inside the second barrier in atm. Thus, the righthand side of Eq. (3) is the pressure-volume product (PV) of the carrier gas remaining within the second barrier in atm cc in the finite time element. The lefthand side of Eq. (3) is from the perfect gas law, NRT=PV; the formulation used in Eq. (3) avoids evaluating N, the number of molecules, or supplying R, the universal gas constant.

Equation (4) is the mass of carrier gas in atm cc that has accumulated within the second barrier in all the preceding finite time elements.

Equation (5) is the pressure in atm within the second barrier due to the carrier gas that was there initially (supplied as the seed value, $sumNRTb_0 = Vb*Pb_0$) plus the carrier gas that accumulated there in all the preceding finite time elements.

Equation (6) through Eq. (8) are the mass balance of the radioactive aerosol particles in the carrier gas. AD is the density of the radioactive aerosol particles in equilibrium with solid (non-aerosol) material. The value of AD is 9×10^{-6} gm/cc of the gas covering the solid plutonium or uranium oxide. AD is also the density of the radioactive aerosol particles in the carrier gas leaking from the first or source barrier.

Equation (6) is the density of the radioactive aerosol particles within and leaking out of the second barrier in gm/cc of carrier gas. The equation sets the aerosol density within and leaking from the second barrier in a finite time element equal to the mass of aerosol particles that has accumulated inside the second barrier in all preceding finite elements divided by the volume of the gas space inside the barrier. The "if" function of Eq. (6) recognizes that initially the aerosol coming from the source barrier is diluted with the gas inside the second barrier, but when the aerosol density inside the second barrier reaches AD, the density of aerosol particles over non-aerosol material, it does not increase further. No consideration is given to the "condensation" of aerosol particles into non-aerosol material within the second barrier, by processes such as settling, or to the "sticking" of radioactive aerosol particles on the surfaces within the second barrier. Because "condensation" and "sticking" of aerosol particles are not considered, the analysis in this paper is conservative; that is, it predicts more radioactive material leakage in a given time than really occurs.

Equation (7) is the mass of radioactive aerosol particles in grams that stay within the second barrier in a finite element. The righthand side of Eq. (7) is the difference between the mass of radioactive aerosol particles leaking in from the first or source barrier minus the mass of radioactive aerosol particles leaking out of the second barrier in a finite element. The righthand side of Eq. (7) is the carrier gas leakage rate of the first and second barriers, La and Lb, in cc/sec times the density of the radioactive aerosol particles in the carrier gas leaking from the first and second barriers, ADa and ADb, in gm/cc multiplied by the size of the finite time element, delt, in sec. Thus the righthand side of Eq. (7) is the mass of radioactive aerosol particles remaining within the second barrier in grams in the finite time element.

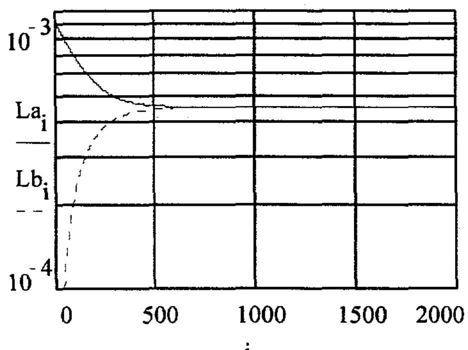
Equation (8) is the mass of radioactive aerosol particles in grams that has accumulated within the second barrier in all preceding finite time elements.

Equation (9) is not part of the mass balance within the second barrier but is added because we want to know the mass of radioactive material that escapes from the second barrier as a function of time.

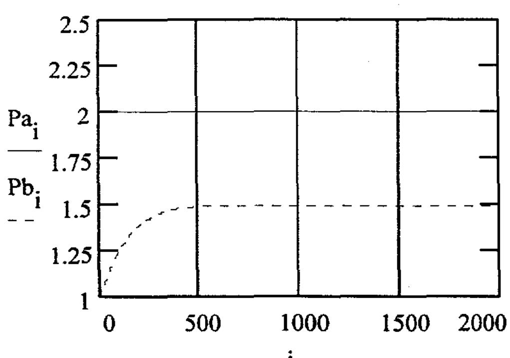
The seed values to start the solution of the matrix are set at values 1×10^{-30} or less. Also in the matrix for three to six barriers, the equations are modified to return values of 1×10^{-40} if the value in the finite element is zero or negative. Although the matrix solution settles into positive values after the first few finite increments and the values at times of interest are unaffected, zero and negative values in the initial finite elements are undesirable because the results display best on semilog graphs.

The time interval, delt, or size of the finite element is set as small as possible and allows the model to approach equilibrium in a reasonable amount of computer time. Testing of the finite element model showed that it is stable with finite elements up to 50,000 secs with six boundaries with a gas leak rate of 1×10^{-3} cc/sec air, a free volume of 1,000 cc inside each barrier downstream of the source barrier. With a finite element larger than 50,000 secs, the model oscillates. The results of the model should always be examined and a smaller finite element used if they oscillate.

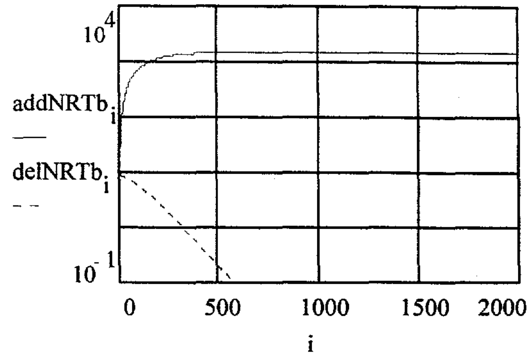
Figures 1 through 6 display the model results for the two boundaries with a free volume of 3,000 cc available inside the second barrier and a finite element of 10,000 sec (~2.8 hours). The results are representative of DOE's 6M configurations, which have about 3,000 cc total free space between the source and 2R but usually have three or more barriers.



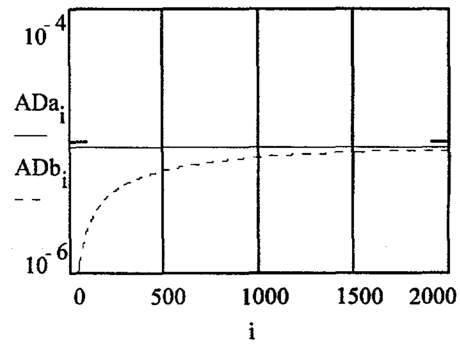
Carrier Gas Leakage Rates, cc/sec
Figure 1



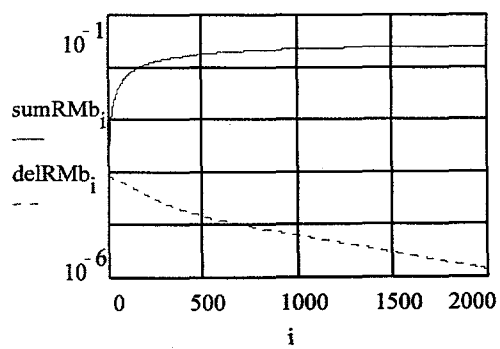
Pressure Inside Boundary, atm
Figure 2



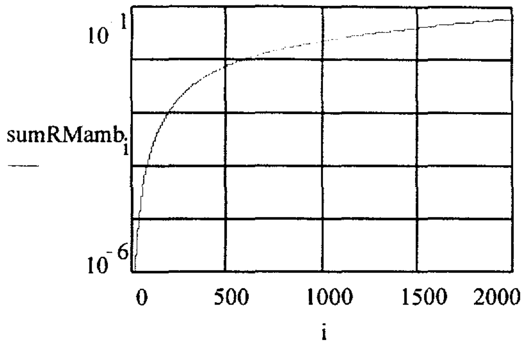
Carrier Gas Accumulation Inside Boundary, cc
Figure 3



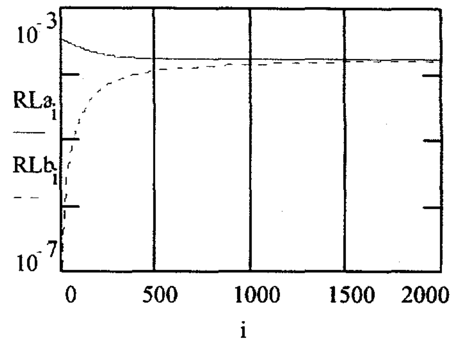
Density of Radioactive Particles In Carrier Gas, gm/cc
Figure 4



Radioactive Particle Accumulation Inside Boundary, gm
Figure 5



Radioactive Particles Escaping To Ambient, gm
Figure 6



Radioactivity Leakage Rate, A_2/hr
Figure 7

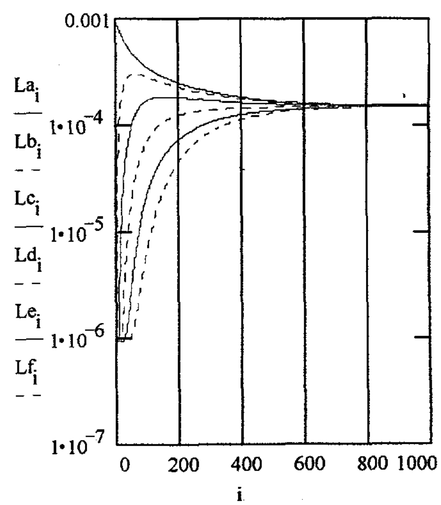
The graphs of each of the nine matrix equations in the preceding figures show that the finite element model behaves as expected. The gas leak rate in cc/sec of the source barrier, La, decreases and the gas leak rate of the second barrier, Lb, increases (Fig. 1) until the pressure within the second barrier, Pa, reaches an equilibrium value of 1.5 atm at about 500 finite elements or about 58 days (Fig. 2). The mass of carrier gas in atm cc accumulating within the second barrier, addNRTb =sumNRTb-Vb*Pb_o, reaches equilibrium in about 500 finite elements.

As expected, the density of the radioactive aerosol particles, ADb, in gm/cc of the carrier gas (Fig. 3) within and leaking from the second barrier is much slower in reaching equilibrium than is the carrier gas. The density of the radioactive aerosol particles in and leaking from the second barrier is close to the equilibrium value of 9×10^{-6} gm/cc at 1,000 finite elements or 116 days.

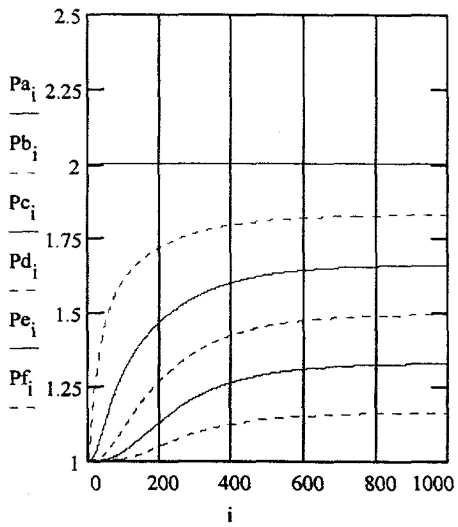
The finite element model shows the magnitude of the two effects of nesting a watertight source with a leak rate of 1×10^{-3} cc/sec air inside an equally leaky second barrier as follows:

1. The gas leak rate of the two barriers decreases from 1×10^{-3} cc/sec air individually to 4.5×10^{-4} cc/sec air in series due to the fact that the leak hole size is not changed by nesting the barriers but the pressure drop from the source to the ambient is divided equally between the two barriers.
2. Radioactivity leakage with two barriers is greatly delayed because of the dilution and mixing in the free space inside the second barrier. In Fig. 7, the radioactivity leakage rate, RLa for the first barrier and RLb for the second barrier, is displayed in transportation terms of A_2/hr for the two barrier 6M containing plutonium oxide that was displayed above. The radioactivity leakage reaches equilibrium at about 1,000 finite elements or 116 days. The radioactivity leakage rate limit of 1×10^{-6} A_2/hr is reached in 22 finite time elements or 2.5 days compared with one barrier being instantaneously 370 times greater than the limit.

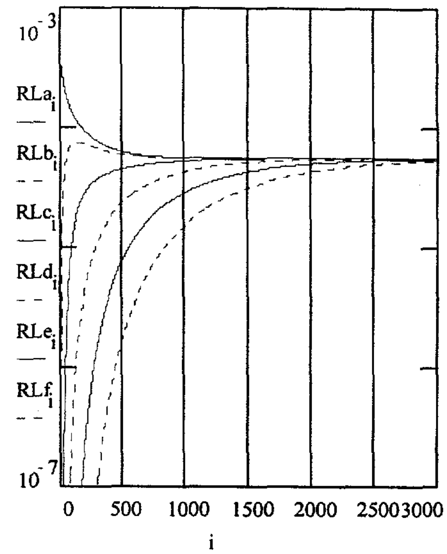
Six Barriers. The finite element model has been run for up to six barriers with the 3,000 cc of free space that is typical of DOE's 6M configurations split equally among the five downstream barriers. No penalty is taken for the free volume taken up by the material of the barriers to compensate for the fact that up to two of the barriers are plastic bags or jars inside the innermost food-pack can. The graphs of the leakage rates of the carrier gas and radioactivity leakage rates in Figs. 8 through 11 show that the finite element model with six barriers behaves just as the two barrier model does.



Carrier Gas Leakage Rates, cc/sec
Figure 8

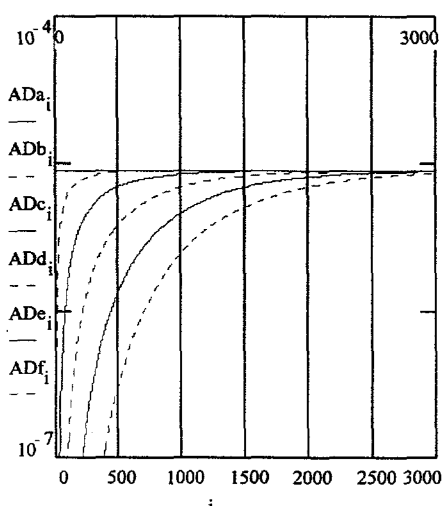


Pressure Inside Boundaries, atm
Figure 9



Radioactivity Leakage Rate, A_2/hr
Figure 11

However, instead of reaching the regulatory limit of 1×10^{-6} A_2/hr in 2.5 days as in the two barrier case, the 6M with six barriers reaches the regulatory limit (Fig. 11) in 52.1 days (450 finite time elements). The following table gives the time to reach the regulatory limit as predicted by the finite element model for two to six barriers.



Density of Radioactive Particles In Carrier Gas, gm/cc
Figure 10

Days to Reach Regulatory Limit on Radioactivity Release Rate, 1×10^{-6} A_2/hr

No. Barriers	F. E. Model
2	2.5
3	11.0
4	22.4
5	35.9
6	52.1

The results for plutonium oxide are encouraging; the calculated delay times with three, four, five, and six barriers are long enough to provide containment of plutonium oxide in the 6M package with watertight food-pack cans, plastic bags, and pipe threaded 2R inner container in shipping plutonium oxide that was not expected from the gas leak rate of the barriers taken individually.

Future Work with the Finite Element Model

The finite element model of nested leaky barriers has been run enough to understand and explain its behavior.

It would be helpful if the effect of plutonium oxide aerosol particles "condensing" to solid material inside each barrier and the effect of "sticking" on the surfaces inside each barrier could be added to the finite element model. Also, the assumption of a constant driving force to cause leakage should be replaced with a more realistic assumption that allows the package to equilibrate with the ambient and then be subjected to an intermittent driving force due to weather and elevation changes.

Food-pack cans and plastic bags are gas bubble leak tested because neither are capable of supporting the pressure difference applied in helium leak testing. In a helium leak test, the food-pack can or plastic bag is placed in a vacuum with 1 atmosphere pressure inside the can or bag. Obviously the plastic bag will burst under these conditions. Neither are food-pack cans strong enough to support 1 atmosphere pressure difference inside to outside; the rolled joint between the lid and can body will be torn open by the distortion of the lid and body. In transportation service, there will not be 1 atmosphere pressure difference across the can wall. Thus, plastic bags and food-pack cans will certainly fail the helium leak test because of the mechanical load the test imposes on them, but they may very well have a leak rate much better than watertight with the mechanical load imposed under actual transportation conditions.

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